Letter

Effects of scattering on the field-induced T_c enhancement in thin superconducting films in a parallel magnetic field

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The problem of the normal-superconducting phase boundary for films in a parallel magnetic field, discussed in the classical paper by Ginzburg and Landau for temperatures close to the critical, is revisited with the help of the microscopic BCS theory for arbitrary temperatures taking pair-breaking and transport scattering into account. Although confirming experimental findings of the T_c enhancement by the magnetic field, we find that the transport scattering pushes the phase transition curve to higher fields and higher temperatures for nearly all practical scattering rates. Still, the T_c enhancement disappears in the dirty limit. We also consider intriguing changes, such as reentrant superconductivity, caused to the phase boundary by pair-breaking magnetic ions spread on one of the film faces. These features await experimental verification.

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I. Introduction. Thin films are major elements of superconducting devices, such as bolometers, superconducting quantum interference devices, fault-current limiters, and qubits for quantum computers. Their physical properties in the superconducting state determine device performance. Unexpected properties of films in the parallel magnetic field have been recorded [1]. The enhancement of the critical temperature T_c above the zero-field T_c in thin superconducting films in parallel fields is now an established experimental fact [2–4]. A few nontrivial mechanisms for this enhancement were suggested [3,5]. However, the opinion expressed in Ref. [2] leans toward early work [6,7], which describes this phenomenon as a consequence of the "bare" classical weak-coupling BCS theory.

The main point of this interpretation is that at the second-order phase transition, the order parameter satisfies a linear equation (as in Helfand and Werthamer treatment of the upper critical field $H_{c2}(T)$ [8])

$$\boldsymbol{\Pi}^2\Delta = k^2\Delta, \tag{1}$$

where $\boldsymbol{\Pi} = \nabla + 2\pi i\boldsymbol{A}/\phi_0$ with the vector potential $\boldsymbol{A}$ and the flux quantum ϕ_0. In fact, this equation holds at any second-order transition from the normal to the superconducting (SC) state away from H_{c2}, e.g., in proximity systems or at H_{c3}, provided $k^2 = -1/\xi^2$ satisfies the self-consistency equation of the theory [7,9]. It turned out that the coherence length ξ, so evaluated depends not only on temperature and scattering, but also on the magnetic field (except in the dirty limit or near T_c). The field dependence has been confirmed in scanning tunneling measurements of the length scale of spatial variation of Δ [10], in muon spin rotation data [11], and in data on macroscopic magnetization $M(H)$ [12] for a number of materials.

Solving Eq. (1), one imposes certain boundary conditions on the order parameter Δ. In the bulk problem of H_{c2}, $\Delta(\boldsymbol{r})$ should be finite everywhere, in the problem of H_{c3} for nucleation of SC at the sample surface one requires $\partial_x\Delta = 0$ at the surface (x is normal to the surface [13]) for a thin film in the parallel field this gradient is required to vanish on both film faces [7].

In this Letter, we extend the earlier treatment [7] by including pair-breaking scattering having in mind possible interpretations for experimental results described in Ref. [2]. Besides, we consider the effect of replacement of the condition $\partial_x\Delta = 0$ at one of the film surfaces with $\Delta = 0$ to describe the pair breaking by magnetic ions spread at this surface (as described in Ref. [2]). The resulting phase transition curves have several surprising and unexpected features, which demonstrate an extreme sensitivity of these curves to the film environment (i.e., to boundary conditions).

Consider an isotropic material with both magnetic and nonmagnetic scatterers; τ_m and τ are the corresponding average scattering times. The problem of the second-order phase transition from the normal to SC phases can be addressed on the basis on Eilenberger quasiclassical version of Gor'kov's equations for normal and anomalous Green's functions g and f. At the second-order phase transition, $g = 1$ and we are left with a linear equation for f [14,15],

$$(2\omega^+ + \boldsymbol{v}\cdot\boldsymbol{\Pi}),\quad f = 2\Delta/\hbar + \langle f\rangle/\tau^-, \tag{2}$$

$$\omega^+ = \omega + \frac{1}{2\tau^+},\quad \frac{1}{\tau^\pm} = \frac{1}{\tau} \pm \frac{1}{\tau_m}. \tag{3}$$

Here, $\boldsymbol{v}$ is the Fermi velocity, $\Delta(\boldsymbol{r})$ is the order parameter; Matsubara frequencies are defined by $\omega = \pi T(2n+1)$ with an integer n; in the following (except some final results) we set $\hbar = k_B = 1$; $\langle\cdots\rangle$ stand for averages over the Fermi surface. Solutions f of Eq. (2) along with Δ should satisfy

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the self-consistency equation,

$$\frac{\Delta}{2\pi T}\ln\frac{T_{c0}}{T}=\sum_{\omega>0}\left(\frac{\Delta}{\omega}-\langle f\rangle\right), \tag{4}$$

where T_{c0} is the critical temperature in the absence of pair-breaking scattering.

Repeating the derivation of Ref. [9], one finds (see the outline in Appendix),

$$\langle f\rangle=\Delta\frac{2\tau^-S}{2\omega^+\tau^- - S}, \tag{5}$$

where S is given by a series,

$$S=\sum_{j,m=0}^{\infty}\frac{(-q^2)^j}{j!(2m+2j+1)}\left(\frac{(m+j)!}{m!}\right)^2\left(\frac{\ell^+}{\beta^+}\right)^{2m+2j}\times\prod_{i=1}^{m}[k^2+(2i-1)q^2],\quad q^2=\frac{2\pi H}{\phi_0}, \tag{6}$$

where,

$$\ell^+=v\tau^+,\quad \beta^+=1+2\omega\tau^+. \tag{7}$$

This sum can be transformed to an integral, which is more amenable for the numerical work [7],

$$S=\sqrt{\frac{\pi}{u}}\int_0^1\frac{d\mu\,(1+\mu^2)^\sigma}{(1-\mu^2)^{\sigma+1}}\left[\text{erfc}\frac{\mu}{\sqrt{u}}-\cos(\pi\sigma)\text{erfc}\frac{1}{\mu\sqrt{u}}\right],\quad u=\left(\frac{q\ell^+}{\beta^+}\right)^2. \tag{8}$$

Introducing dimensionless quantities,

$$h=H\frac{2\pi d^2}{\phi_0}=q^2d^2,\quad P^\pm=\frac{\hbar}{2\pi T_{c0}\tau^\pm}=P\pm P_m, \tag{9}$$

and the reduced thickness,

$$D=d\frac{2\pi T_{c0}}{\hbar v}, \tag{10}$$

one obtains

$$u=\frac{h}{D^2[P^+ + t(2n+1)]^2}. \tag{11}$$

The parameter σ as defined in Ref. [7] is

$$\sigma=\frac{1}{2}\left(\frac{k^2}{q^2}-1\right). \tag{12}$$

This parameter depends on the phase transition in question: It is easy to see that $\sigma=-1$ at $H_{c2}(T)$. For H_{c3} near T_c of a half-space sample the result of Saint-James and DeGennes causes $\sigma=-0.795$ [13]; transport scattering leads to the temperature dependence of σ [16,17].

For numerical work we recast the self-consistency relation (4) combined with Eq. (5) to dimensionless form

$$-\ln t=\sum_{n=0}^{\infty}\left[\frac{1}{n+1/2}-\frac{2tS}{2t(n+1/2)+P^+ - SP^-}\right], \tag{13}$$

with the reduced temperature $t=T/T_{c0}$.

II. Symmetric boundary conditions $\Delta'(\pm d/2)=0$. As mentioned above, the order parameter at a second-order phase transition satisfies $\Pi^2\Delta=k^2\Delta$. Choose the plane (y,z) parallel to the film and $x=0$ in the film middle. Denoting,

$$s=qx,\quad \eta=-k^2/q^2, \tag{14}$$

we obtain a differential equation,

$$\Delta''(s)-s^2\Delta(s)=-\eta\Delta(s), \tag{15}$$

so $-\eta$ is the eigenvalue of the linear operator at the left-hand side. The general solution is as follows:

$$\Delta=e^{-s^2/2}\left[{}_1F_1\left(\frac{1-\eta}{4},\frac{1}{2},s^2\right)+Cs\,{}_1F_1\left(\frac{3-\eta}{4},\frac{3}{2},s^2\right)\right], \tag{16}$$

where ${}_1F_1$'s are confluent hypergeometric functions and C is an arbitrary constant. The symmetry with respect to the film middle gives $C=0$, and the condition $\Delta'(\pm d/2)=0$ yields

$$(1-\eta)\,{}_1F_1\left(\frac{5-\eta}{4},\frac{3}{2},\frac{h}{4}\right)={}_1F_1\left(\frac{1-\eta}{4},\frac{1}{2},\frac{h}{4}\right). \tag{17}$$

Hence, for a given field h, the eigenvalue η can take only a certain value, the root of this equation.

Given $\eta(h)$, we evaluate $\sigma=(k^2/q^2-1)/2=-(\eta+1)/2$ for this value of h, and, therefore, we can calculate $S(u,\sigma)=S(h,t,n)$ for given P, P_m, and D and solve the self-consistency equation for t at this h. Scanning h we recover the whole transition curve curve $t(h)$. In general, scanning t would not work because $h(t)$ might happen to be multivalued.

The numerical results for purely transport scattering are shown in Fig. 1. It is worth noting that the transport scattering causes an increase of the T_c enhancement up to $P\sim 10$ and only for strong scattering with $P>10$ it suppresses the effect in agreement with the general theoretical statement that the effect should disappear in the dirty limit [7,9].

The upper panel of Fig. 2 shows phase boundaries for the clean case and a set of thicknesses from $D=0.1$ to $D=5$ at temperatures close to T_{c0}. The remarkable feature here is that for $D\lesssim 2.8$ these boundaries deviate from T_{c0} with increasing parallel field H in a "wrong" direction that results in enhancements of $T_c(H)$. The phase boundary becomes standard for thicker films with $D\gtrsim 2.8$ (the Ginzburg-Landau (GL) theory predicts this value to be 2.6 [7]).

The lower panel shows that at T close to zero, the field,

$$H=\frac{h}{D^2}\frac{2\pi\phi_0T_{c0}^2}{\hbar^2v^2} \tag{18}$$

(in common units) is nearly constant for small thicknesses in the interval $0.1<D\lesssim 1$, but it decreases for thicker films. One should have in mind that the "laminar" structure of $\Delta(x)$ of thin films with increasing thickness becomes unstable and gives way to vortices with $\Delta(x,y)$ as had been shown in Refs. [18,19] within GL theory. Although the large values of D are irrelevant for the film problem, it is interesting to note that formally $H(0)$ for large D is close to $H_{c2}(0)$.

Figure 3 shows that the magnetic scattering, whereas strongly suppressing the critical temperature, leaves the T_c enhancement effect, i.e., (t^*-t_c) nearly unchanged. For curves

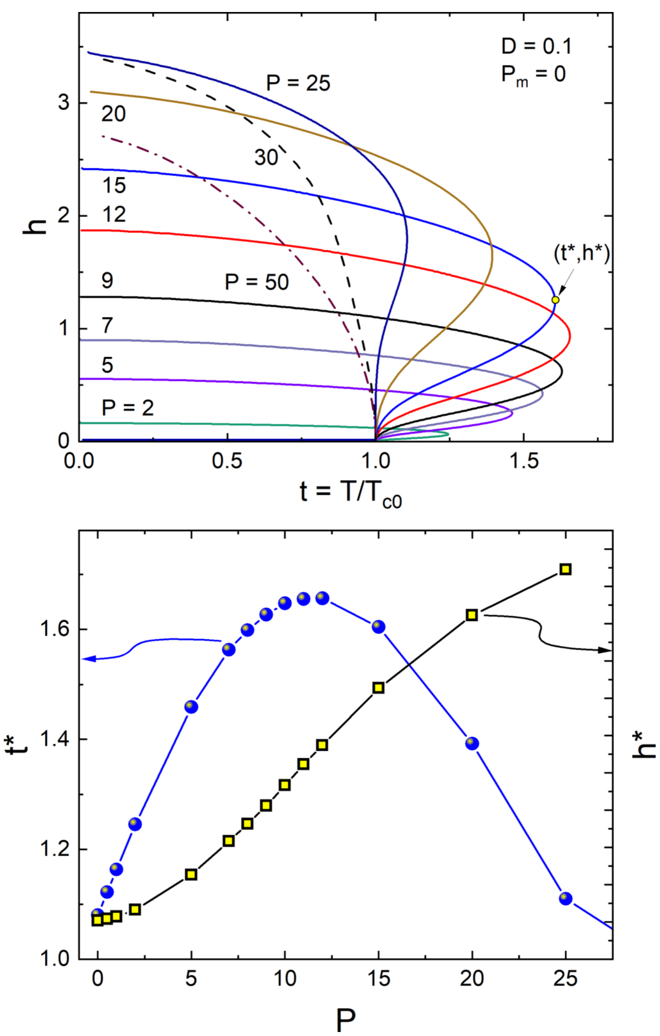

FIG. 1. The upper panel: the phase boundary $h(t)$ for a set of different transport scattering rates P indicated. The position of the maximum enhancement (t^*, h^*) is shown for one curve, $P = 15$. The lower panel: the position of the maximum enhancement t^* (left axis) and h^* (right axis) as a function of P. On both panels $D = 0.1$, $P_m = 0$.

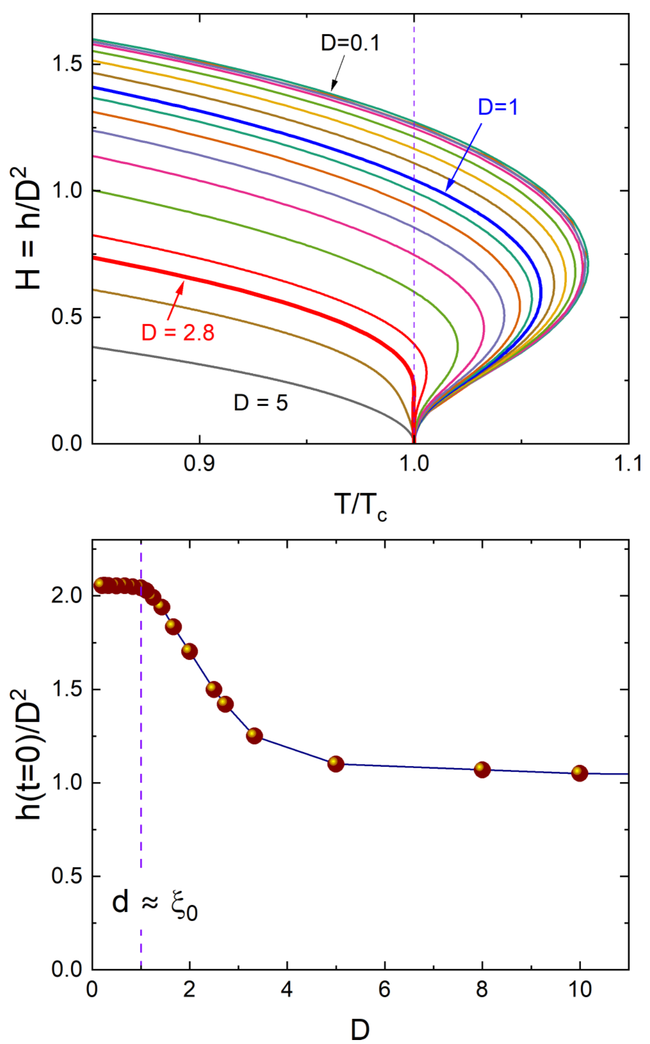

FIG. 2. The upper panel: the clean-limit ($P = P_m = 0$) phase boundary $h(t)$ zoomed at high temperatures for a set of thicknesses indicated. The T_c enhancement disappears at approximately $D = 2.8$. The lower panel: the close-to-zero-temperature field $h(0)/D^2 \propto H(0)$ vs D.

$P_m = 0$ and $P_m = 0.13$, T_c drops by a factor of 5, whereas $(t^* - t_c)$ changes by about 30%.

It should be noted that the scattering parameters P and P_m refer to the bulk properties of the film material. Effects of magnetic ions on one of the film faces can be taken into account by the boundary conditions at the surfaces rather than by the value of P_m, the subject of the next section.

III. Mixed boundary conditions: $\Delta'(d/2) = 0$ *on one surface and* $\Delta(-d/2) = 0$ *on the other*. These conditions can be realized in a film on an insulating substrate ($\Delta' = 0$) with magnetic ions spread at the other surface ($\Delta = 0$). We have chosen these boundary conditions to demonstrate how sensitive the phase boundary in films placed in the parallel field is to the film environment. In fact, Saint-James and de Gennes pointed this out in their seminal work [13].

The boundary conditions of the section title suffice to determine both the arbitrary constant C and the parameter η of the general solution (16). $\Delta(-d/2) = 0$ yields

$$C = \frac{2}{\sqrt{h}}\, {}_1F_1\left(\frac{1-\eta}{4}, \frac{1}{2}, \frac{h}{4}\right) \Big/ \, {}_1F_1\left(\frac{3-\eta}{4}, \frac{3}{2}, \frac{h}{4}\right). \quad (19)$$

The condition $\partial_x \Delta(d/2) = 0$ results in,

$$3C\left(1+\frac{h}{4}\right) {}_1F_1\left(\frac{3-\eta}{4}, \frac{3}{2}, \frac{h}{4}\right) + \frac{\sqrt{h}}{2}\Bigg[3\, {}_1F_1\left(\frac{1-\eta}{4}, \frac{1}{2}, \frac{h}{4}\right)$$
$$- 3(1+\eta)\, {}_1F_1\left(\frac{1-\eta}{4}, \frac{3}{2}, \frac{h}{4}\right)$$
$$- C\frac{\sqrt{h}}{2}(3+\eta)\, {}_1F_1\left(\frac{3-\eta}{4}, \frac{5}{2}, \frac{h}{4}\right)\Bigg] = 0. \quad (20)$$

At a given h, Eqs. (19) and (20) can be solved for η numerically [η is needed for the power $\sigma = -(\eta+1)/2$ in the integral S].

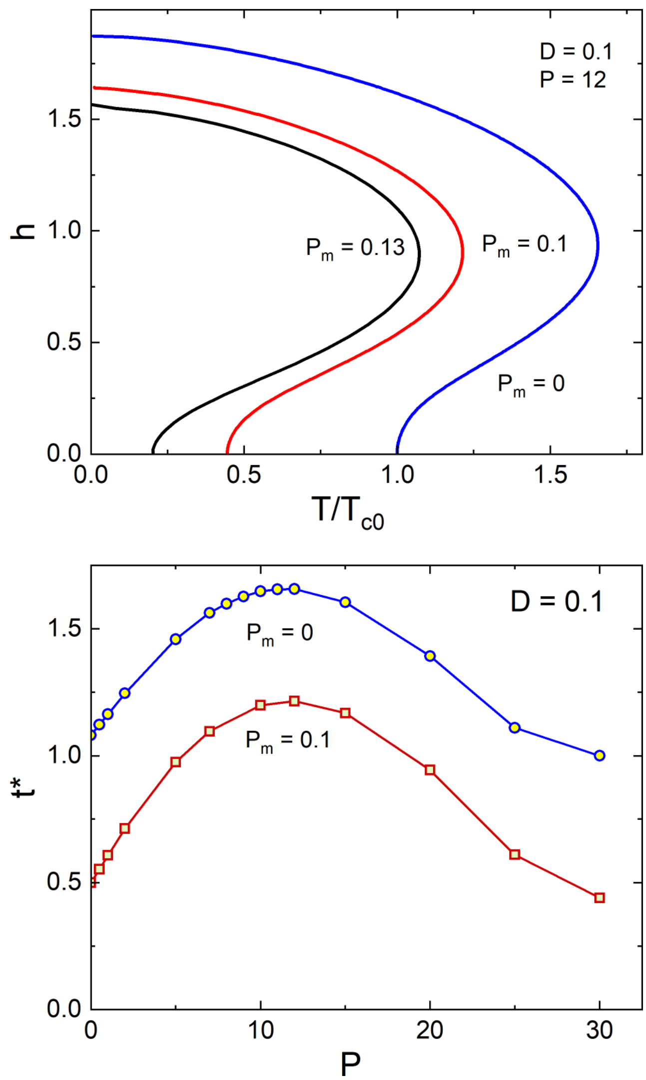

FIG. 3. The upper panel: the phase boundary $h(t)$ for different magnetic scattering rates P_m at a fixed $P = 12$. The lower panel: the maximum t_c enhancement, t^* vs transport scattering rate P for two fixed P_m. At both panels $D = 0.1$.

A. Zero-field $T_c(d)$. The condition $\Delta(-d/2) = 0$ suppresses the film T_c even in the field absence. Evaluation of this suppression is necessary to interpret various transition curves $h(t)$. In zero field, the order parameter at the phase boundary satisfies $\Delta'' = k^2\Delta$ with the solution $\Delta = \Delta_0 \sin|k|(x + d/2)$ with $|k| = \pi/2d$. Furthermore, T_c should be found from the self-consistency Eq. (4) which contains the quantity S via Eq. (5). The shortest way to get S for $H = 0$ is to go to the definition of S as a power series (6) and set in it $q^2 = 2\pi H/\phi_0 = 0$ [9,17],

$$S = \frac{1}{\mu}\tan^{-1}\mu, \quad \mu = \frac{\pi}{2D}\frac{\ell^+}{\beta^+}. \tag{21}$$

The self-consistency equation now reads

$$-\ln t_c = \sum_{n=0}^{\infty}\left[\frac{1}{n+1/2} - \frac{2t_c S}{t_c(2n+1) + P^+ - SP^-}\right], \tag{22}$$

where $t_c = T_c/T_{c0}$ and T_{c0} is the critical temperature of the bulk material in the absence of pair-breaking scattering. The dimensionless parameter μ of Eq. (21) is

$$\mu = \frac{\pi}{2D}\frac{1}{t_c(2n+1) + P^+}, \quad D = d\frac{2\pi T_{c0}}{\hbar v}, \tag{23}$$

so that one can solve the self-consistency equation for $t_c(D)$ numerically.

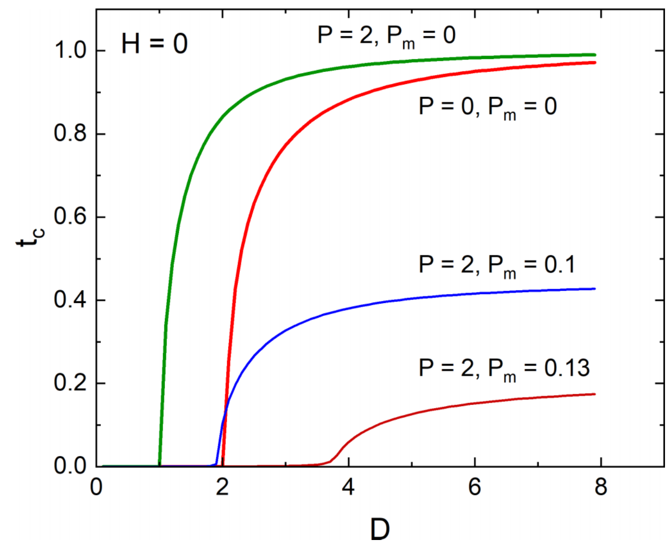

FIG. 4. Zero-field critical temperature $t_c(D)$ as a function of thickness D for the mixed boundary conditions and various combinations of the scattering parameters indicated.

As Fig. 4 shows, the requirement $\Delta = 0$ at one of the film surfaces leads to a progressive reduction of T_c with decreasing thickness. Moreover, T_c turns zero at $D = D_c = 2$ in the clean case, so that in zero field SC is completely suppressed for $D < D_c$. With increasing transport scattering, the sharp break of $t_c(D)$ at D_c moves to smaller thicknesses. Hence, the phenomenology of SC films in parallel fields is quite peculiar. The pair-breaking scattering smears the sharp break in $t_c(D)$ to a smooth crossover, whose position is shifted to thicker films. Our example of $P_m = 0.13$ corresponds to a strong pair breaking and the gapless situation in the bulk (recall that the critical value of P_m where the bulk $T_c(P_m) = 0$ is $P_m = 0.14$, see, e.g., Ref. [15]).

It is instructive to observe that if $D \gg 1$, the parameter μ is small, and Eq. (13) is reduced to the Abrikosov-Gor'kov bulk relation for $t_c(P_m)$,

$$-\ln t_c = \psi\left(\frac{1}{2} + \frac{P_m}{t_c}\right) - \psi\left(\frac{1}{2}\right). \tag{24}$$

B. Phase boundary. For thicknesses substantially exceeding D_c ($D_c = 2$ for the clean limit) the transition curve is of the type $H \sim \sqrt{T_c - T}$ with zero enhancement as shown in Fig. 5.

When $D < D_c = 2$, $T_c = 0$ for $H = 0$. Therefore, the low end of the transition curve should start at $t = 0$ as is seen in Fig. 5 for clean films with $D = 1.8$ and 2. The equilibrium SC does not exist at all out of patches bound by these curves. For SC to exist one should apply field within the area inside these patches. In particular, such a phase boundary implies that the magnetoresistance at a fixed temperature $t \lesssim 0.36$ should have a minimum for $D = 2$. These examples demonstrate that thin films in parallel fields complement the list of phenomena

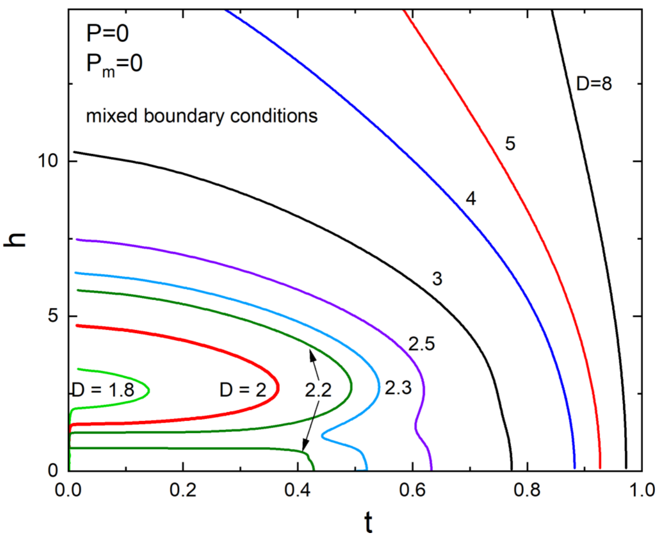

FIG. 5. The phase boundary $h(t)$ for the mixed boundary conditions on the order parameter, $\Delta'(D/2)=0$ on one surface, and $\Delta(-D/2)=0$ on the other in a clean limit for $P=0$, $P_m=0$. The reduced field is $h=(2\pi d^2/\phi_0)H$ and the reduced temperature $t=T/T_{c0}$. The numbers by the curves are dimensionless thicknesses $D=(2\pi T_{c0}/\hbar v)d$.

where the magnetic field "helps" SC instead of suppressing it, see, e.g., Ref. [5].

Unlike the situation with $D<D_c=2$ for $D>D_c$ the zero-field T_c is not zero, therefore, the low end of the transition curve starts at $T_c>0$, examples are shown in Fig. 5 for $D=2.2$ and larger. In the region of thicknesses adjacent to D_c, the phase boundary may take a nontrivial shape shown for $D=2.2,\ 2.3,$ and 2.5 where the transition curve $h(t)$ becomes multivalued.

IV. Summary. We have considered thin superconducting films in a parallel magnetic field. Near T_c, this problem has been discussed in the classic GL paper [20]. Our approach is based on the quasiclassical microscopic theory of Eilenberger [14] that holds for any T.

We have shown that in the presence of impurities, the magnetic ones included, the model [7,9] developed in the late 1980s still works. We obtained conditions for the remarkable effect recently observed [2], the enhancement of the in-field critical temperature $T_c(H)$ above the zero-field $T_c(0)$.

The formal procedure we employ is the same as that used for evaluating the surface critical field H_{c3} for the half-space superconductor [13,16]. The film is considered as two surfaces separated by the film thickness d on the order of coherence length ξ. Then, the boundary conditions for the order parameter $\Delta(x)$ impose solutions, Eq. (16), different from those of Saint-Games and De Gennes. The new element in our approach, absent in that of Ref. [13] near T_c, is that the coherence length $\xi(T)$ at arbitrary temperatures calculated within the BCS (Eilenberger) theory turns out to depend on the magnetic field. Thus, the enhancement of the phase boundary for a film in parallel field to T's larger than the zero field T_{c0} as well as to H larger than the bulk H_{c2} has the same nature as the superconductivity in fields $H_{c2}<H<H_{c3}$ of Saint-Games and De Gennes. In other words, to interpret this enhancement one does not need an extra mechanism not included in classical BCS.

We find that transport scattering amplifies this effect if the scattering parameter $P\sim\xi_0/\ell\lesssim 10$ (ℓ is the mean free path and ξ_0 is the BCS zero-T coherence length). For larger P, the T_c enhancement is suppressed to disappear in the dirty limit [7,9]. This new insight clarifies the role of the transport scattering bringing it in line with the general theory prediction of absent T_c enhancement in the dirty limit. On the other hand, this improves the chances to observe the effect [2,3] because the transport scattering in thin films is usually strong. For example, the experiment [2] registered T_c enhancements in amorphous Pb films with the estimated mean free path $\ell\approx 1$ nm whereas estimates of ξ_0 in Pb range between $\approx$230 and 300 nm. In general, whereas in many modern superconductors, such as high-T_c cuprates or is iron pnictides, ξ_0 is so short that makes it challenging to make such films, in technologically—important Nb it is quite feasible with $\xi_0\approx$ 40 nm.

It should be noted that the effect of magnetic impurities *per se* on the T_c enhancement turned out relatively weak, the standard T_c suppression notwithstanding, see Fig. 3.

The properties of thin films in a parallel magnetic field are very sensitive to physical conditions on their faces. If, for example, pair-breaking magnetic ions are spread over one of the faces whereas the opposite face is on an insulating substrate, then more realistic boundary conditions for the order parameter would be $\Delta=0$ on one face and $\partial_x\Delta(x)=0$ at the other. This arrangement was, in fact, tested in experiment [2].

The major consequence of the condition $\Delta=0$ on one of the faces is that it makes the critical temperature thickness dependent. We find that for $P=P_m=0$, the surface pair breaking kills the SC in zero-field for thicknesses $D<2$ (in units $\hbar v/2\pi T_{c0}$). But as Fig. 5 shows, even for $D<2$, application of the magnetic field may cause the SC *reentrance* at a finite field interval. Hence, the film magnetoresistance at $T=$ const should have a dip in this interval.

For $2<D\lesssim 3$, the competition of reentrance and finite T_c causes the phase transition curve to acquire a nontrivial shape so that $h(t)$ becomes multivalued, see Fig. 5.

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APPENDIX: THE SUM *S* IN THE PRESENCE OF MAGNETIC IMPURITIES

The solution f of Eq. (2) can be written as

$$f=(2\omega^++\boldsymbol{v}\boldsymbol{\Pi})^{-1}(F/\tau^-+2\Delta)$$
$$=\int_0^\infty d\rho\, e^{-\rho(2\omega^++\boldsymbol{v}\boldsymbol{\Pi})}(F/\tau^-+2\Delta). \quad \text{(A1)}$$

Taking the Fermi-surface average we get

$$F = \frac{1}{\tau^-}\int_0^\infty d\rho\, e^{-2\omega^+\rho}\langle e^{-\rho \boldsymbol{v}\boldsymbol{\Pi}}\rangle(F + 2\Delta\tau^-). \tag{A2}$$

The term $\langle\cdots\rangle$ does not contain the scattering parameters, hence, it is the same as that calculated in Refs. [6,7,9] for the clean case,

$$\langle e^{-\rho \boldsymbol{v}\boldsymbol{\Pi}}\tilde{F}\rangle = \sum_{m,j}\frac{(-q^2)^j}{(m!)^2 j!}\frac{(2\mu)!!}{(2\mu+1)!!}\left(\frac{\rho v}{2}\right)^{2\mu}\Pi^{+^m}\Pi^{-^m}\tilde{F}. \tag{A3}$$

Here $\tilde{F} = F + 2\Delta\tau^-$, $\mu = m + j$, and $\Pi^\pm = \Pi_x \pm i\Pi_y$. After integrating over ρ, one obtains from Eq. (A2),

$$F = \frac{1}{2\omega^+\tau^-}\sum_{m,j}\frac{(-q^2)^j}{j!(2\mu+1)}\left(\frac{\mu!}{m!}\right)^2\left(\frac{\ell^+}{\beta^+}\right)^{2\mu}\Pi^{+^m}\Pi^{-^m}\tilde{F}$$

$$\ell^+ = v\tau^+,\quad \beta^+ = 1 + 2\omega\tau^+. \tag{A4}$$

One can check that if no magnetic impurities are involved, this reduces to Eq. (12) of Ref. [9]. Using commutation properties of operators $\Pi^\pm$ in uniform field, one manipulates

$$\Pi^{+^m}\Pi^{-^m}\tilde{F} = \tilde{F}\prod_{i=1}^m[k^2 + (2i-1)q^2] \tag{A5}$$

and obtains

$$F = \Delta\frac{2\tau^- S}{2\omega^+\tau^- - S}, \tag{A6}$$

with

$$S = \sum_{m,j}\frac{(-q^2)^j}{j!(2\mu+1)}\left(\frac{\mu!}{m!}\right)^2\left(\frac{\ell^+}{\beta^+}\right)^{2\mu}\prod_{i=1}^m[k^2 + (2i-1)q^2]. \tag{A7}$$

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